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Neutron Capture Cross Sections and Gamma Emission Spectra from Neutron Capture on $^{234,236,238}\text{U}$ measured with DANCE

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A new measurement of the $^{238}\text{U}(n,\gamma)$ cross section using a thin 48 mg/cm² target was made using the DANCE detector at LANSCE over the energy range from 10 eV to 500 keV. The results confirm earlier measurements. Measurements of the gamma-ray emission spectra were also made for $^{238}\text{U}(n,\gamma)$ as well as $^{234,236}\text{U}(n,\gamma)$. These measurements help to constrain the radiative strength function used in the cross-section calculations.

I. INTRODUCTION

A precise knowledge of the $^{238}\text{U}(n,\gamma)$ cross section is important for many applications ranging from defense and safeguards to nuclear power reactors. There have been many measurements made over the years that are documented in the EXFOR data library [1]. Early evaluations of the data tended to favor cross sections in the keV range that were considered to be too high to be consistent with a careful analysis of power reactor parameters. Subsequent reanalysis of the data and the use of new evaluation techniques resulted in lower cross sections that were in better agreement with expectations [2],[3]. There have been few recent measurements of $^{238}\text{U}(n,\gamma)$, and changes in the evaluated data have been largely due to reanalysis of existing data or new methods for evaluation.

In this paper, we present the results of a new measurement with a thin (48 mg/cm²) target using the Detector for Advanced Neutron Capture Experiments (DANCE) at the Manuel J. Lujan, Jr., Neutron Scattering Center at Los Alamos. By measuring the full energy of the capture cascade, DANCE provides a better control of external backgrounds and backgrounds due to neutron scattering. The high efficiency of DANCE combined with the high neutron flux at LANSCE enabled the use of a thin target to minimize self-attenuation and multiple-scatter corrections. The gamma-ray emission spectrum was also measured, and the two-step cascade distribution was used to

provide additional constraints on the radiative strength function used to calculate the capture cross section.

II. THE DANCE DETECTOR

DANCE is a 4π calorimetric gamma-ray detector that views the “upper tier” water moderator at the Lujan Center spallation neutron source at LANSCE. It consists of 160 BaF₂ crystals, each about 0.751 l in volume and 15 cm deep. The high segmentation permits an analysis of the gamma-ray multiplicity as well as maintaining a reasonable count rate in each crystal when studying radioactive targets. The face of each crystal is 17 cm from the sample center. The sample was located 20.25 m from the moderator, and the beam spot at the sample location was about 1 cm in diameter. The sample was surrounded by a 6 cm thick ⁶LiH sphere to attenuate neutrons scattered from the sample.

The DANCE data acquisition system consists of two 8-bit fast transient digitizers for each crystal. Each digitizer has a 2 ns channel width, and 128 kilochannels of memory. For this experiment, the two digitizers were triggered consecutively for a total of 500 μsec acquisition time. More information on the acquisition system is in ref. [4].

Three neutron monitors located about 2 m downstream of the sample location were used to measure the beam intensity. The monitors were a ²³⁵U fission chamber, a BF₃ counter, and a ⁶Li(n,t) monitor consisting of a thin ⁶LiF deposit in the beam and a Si surface-barrier detector at 90 degrees to the beam. The measured beam flux, for an average proton beam current of 96.9 μA , is shown in Fig.

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1. The ${}^6\text{Li}$ monitor was used for neutron energies less than 10 keV and the ${}^{235}\text{U}$ monitor for energies greater than 10 keV. The two were normalized to each other over the range 3 to 10 keV. The large dips in the flux at 34.8, 86.2, and 143 keV are due to resonances in the structural Al which was used for the moderator and various beam-line windows. Data in the vicinity of these resonances was excluded because the timing resolution of the monitors and the DANCE BaF_2 was different, leading to a problematic correction.

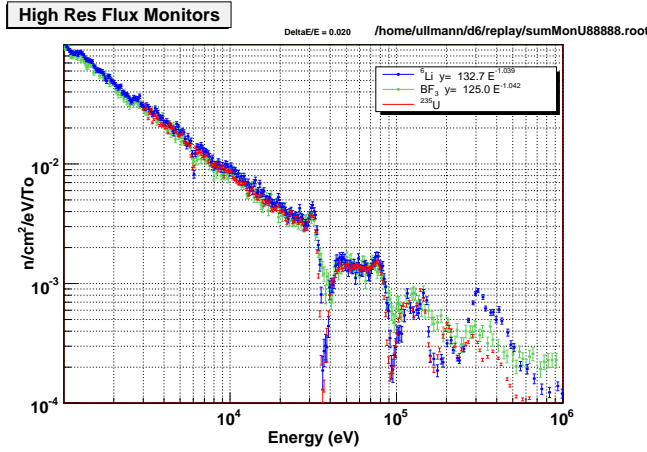


FIG. 1. Neutron flux measured at the monitor location for the three neutron monitors. The average proton beam current was 96.9 μA .

III. ${}^{238}\text{U}(n, \gamma)$ CROSS SECTION

The measured ${}^{238}\text{U}(n, \gamma)$ cross section from 10 keV to 700 keV is shown in Fig 2. The cross section was normalized to the area of weak resonances at 80.74, 145.60, and 165.30 eV. The area for these resonances was calculated using the procedures in the code SAMMY [5] for resolution broadening, self-attenuation, and multiple scatter correction. The multiple-scatter correction for these resonances was less than 1%. The data is binned in $dE/E = 0.05$ energy bins. A comparison is made to selected measurements from the EXFOR data base. Also shown is the ENDF/B-VII.0 evaluation calculation made by Kawano using the code CoH, and reported in ref. [6]. This calculation used optical model parameters from Sukhovitskij [7], level densities from Kawano [8], and a “Generalized Lorentzian” E1 radiative strength function [9]. The calculated cross section was normalized to the average value of Γ_γ . The calculation required $\langle \Gamma_\gamma \rangle = 17$ meV to reproduce the data, compared to the tabulated value of 23.6 meV [10]. This trend of needing lower $\langle \Gamma_\gamma \rangle$ values than measured has been observed in calculations for other actinides measured at DANCE. This difference is not understood.

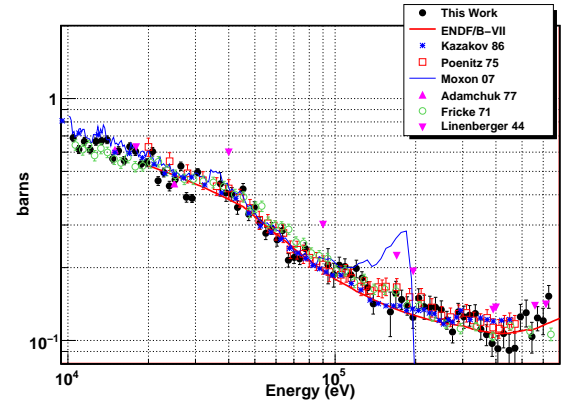


FIG. 2. Cross section for ${}^{238}\text{U}(n, \gamma)$ measured over the energy range 10 to 700 keV. Also shown are selected measurements from EXFOR [1]. The high-resolution data of Moxon is represented as a continuous line. Also shown is the ENDF/B-VII evaluation.

IV. GAMMA-RAY SPECTRA

An additional test of the radiative strength functions and level densities can be made by comparing the measured gamma-ray energy distribution, for a given gamma-ray multiplicity, with calculated values. To do this, simulated gamma-ray cascades were generated using the DICEBOX Monte-Carlo code [11] and propagated through a GEANT4 model of the DANCE detector [12]. The GEANT simulation models the detector geometry, scintillator resolution and thresholds, and gamma-ray interactions to simulate the observed spectra. The average of ten “realizations” of the spectra was then compared to the measurement. This comparison is largely qualitative.

We chose to study the “two-step” gamma spectra from well-resolved low-lying resonances in ${}^{234,236,238}\text{U}(n, \gamma)$, and used data taken using “thin” (approximately 1 to 2 mg/cm^2) targets. These targets were made by electroplating the target material on thin Ti backing foils. The data were gated on the resonance energy and by a window of $S_n \pm 0.5$ MeV on the summed-energy spectrum. Backgrounds determined with the same windows were subtracted from the data.

DICEBOX calculations were made using an E1 Generalized Lorentzian [9] form for the radiative strength function with parameters determined from ref [13]. The level density was a back-shifted Fermi gas form with parameters determined by von Egidy [14]. The calculations compared to the data are shown in Figures 3, 4, and 5. Also shown for ${}^{238}\text{U}$ are calculations using the Generalize Lorentzian radiative strength function made using the code CGM. It can be seen that the calculations using a pure E1 GL strength function generally do a poor job of representing the data.

It has been suggested that additional strength at low excitation energies, where the level density is the highest and the shape of the E1 strength function is not well de-

terminated, is needed to represent the data. To test this, DICEBOX calculations with low-lying M1 strength were made. This strength was represented by two Lorentzians, one at $E_R=2.0$ MeV, $\Gamma_R=0.6$ MeV, $\sigma_R=1.3$ mb, representing the scissors mode, and another at $E_R=7.0$ MeV, $\Gamma_R=3.0$ MeV, $\sigma_R=4.7$ mb, representing the “spin-flip” M1 mode. These parameters were estimated from systematics but adjusted to reproduce the ^{238}U data. The same parameters were also used for the $^{234,236}\text{U}$ calculations. The ^{238}U data are well-represented by the inclusion of the low-lying M1 strength, the general features of the ^{236}U are represented, but the calculation for ^{234}U is not optimal.

While these calculations used general systematics to suggest the M1 strength, there has been a great deal of quantitative experimental and theoretical work on low-lying M1 strength in nuclei [15]. In the rare-earth nuclei, this work suggests that while the scissors-mode strength in odd-mass nuclei is somewhat fragmented compared to adjacent even-mass isotopes, the total strength determined from even-mass systematics is present [16]. However, while there has been considerable experimental and theoretical work on the U isotopes [17–20] and other actinides [21], the situation is not as clear. Careful calculations based on the quantitative information are required.

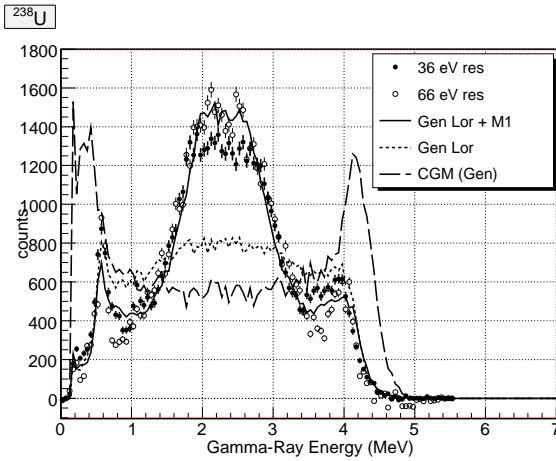


FIG. 3. Two-step cascade gamma-ray spectrum from the 36 eV and 66 eV resonances in $^{238}\text{U}(n, \gamma)$. Calculations made using the DICEBOX code with an E1 Generalized Lorentzian (GL) strength function and an GL + M1 strength function are shown. $E_{crit}=0.83$ MeV, $S_N=4.81$ MeV. Also shown are calculations made with the CGM code using a GL strength function. All spectra are normalized to the 36 eV resonance.

V. CONCLUSIONS

A new measurement of the ^{238}U neutron capture cross section, using a 48 mg/cm² target, confirms previous experimental results. Hauser-Feshbach calculations of the cross section reproduce the energy dependence of the data, but require an $\langle \Gamma_\gamma \rangle$ normalization that is not

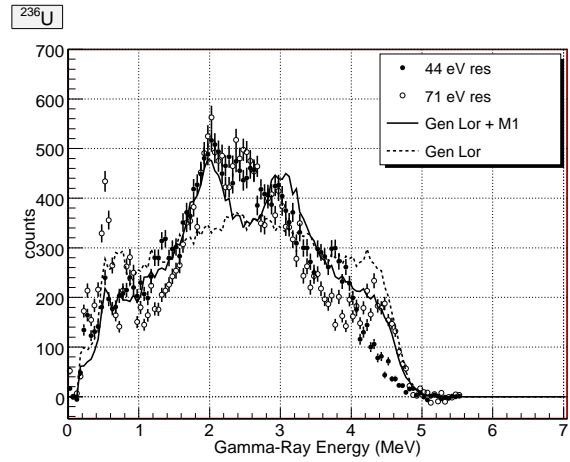


FIG. 4. Two-step cascade gamma-ray spectrum from the 44 eV and 71 eV resonances in $^{236}\text{U}(n, \gamma)$. Calculations made using the DICEBOX code with an E1 Generalized Lorentzian (GL) strength function and an GL + M1 strength function are shown. $E_{crit}=0.60$ MeV, $S_N=5.13$ MeV. All spectra are normalized to the 44 eV resonance.

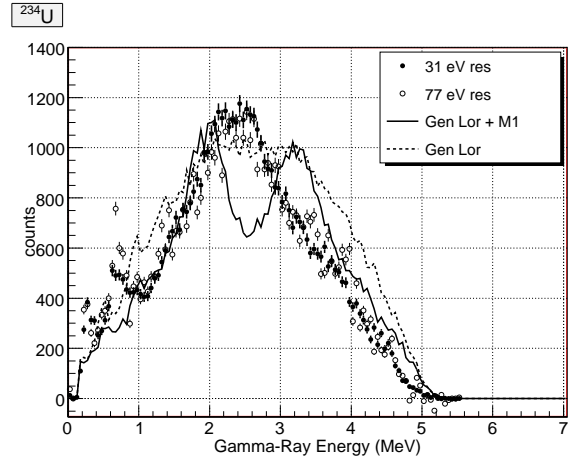


FIG. 5. Two-step cascade gamma-ray spectrum from the 31 eV and 77 eV resonances in $^{234}\text{U}(n, \gamma)$. Calculations made using the DICEBOX code with an E1 Generalized Lorentzian (GL) strength function and an GL + M1 strength function are shown. $E_{crit}=0.50$ MeV, $S_N=5.30$ MeV. All spectra are normalized to the 31 eV resonance.

consistent with the measured value. The gamma-ray spectrum corresponding to two-step cascades has been measured for low-lying resonances in $^{234,236,238}\text{U}(n, \gamma)$. Calculations of the shape of the gamma-ray spectrum using only an E1 Generalized Lorentzian radiative strength function do not adequately represent the data, however the addition of low-lying M1 strength in the calculation improves the representation. Further calculations based on quantitative strengths measured in complementary reactions or determined theoretically are needed to consistently describe the observations. This work is in progress.

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- [1] The EXFOR tabulation of data and references is available through the National Nuclear Data Center at www.nndc.bnl.gov.
 - [2] T. Kawano, *et al.*, PROC. INT. CONF ON NUCL. DATA Gatlinburg, (1994).
 - [3] Y. Kanda, *et al.*, NEA-WPEC-4.
 - [4] J.M. Wouters, *et al.*, IEEE TRANSACTIONS ON NUCLEAR SCIENCE **53**, 880 (2006).
 - [5] N.M. Larson, ORNL/TM-9179/R7, Oak Ridge National Laboratory, Oak Ridge TN, USA (2006). Also available as ENDF-364/R1.
 - [6] P. Young, *et al.*, NUCL. DATA SHEETS **108**, 2589 (2007).
 - [7] E.S. Soukhovitskij, *et al.*, J. NUCL. SCI. TECH. **37**, 120 (2000).
 - [8] T. Kawano, *et al.*, J. NUCL. SCI. TECH. **43**, 1 (2006).
 - [9] J. Kopecky and M. Uhl, PHYS. REV. C**42**, 1941 (1990).
 - [10] Reference Input Parameter Library: www-bds.iaea.org/ripl2.
 - [11] F. Becvar, NUCL. INSTRUM. METH. A **417**, 434, (1998).
 - [12] M. Jandel, *et al.*, NUCL. INSTRUM. METH. B **261**, 117 (2007).
 - [13] S.S. Dietrich and B.L. Berman, AT. DATA AND NUCL. DATA TAB. **38**, 199 (1988).
 - [14] T. von Egidy and D. Bucurescu, PHYS. REV. C**80**, 054310 (2009).
 - [15] K. Heyde, P. von Neumann-Cosel, and A. Richter, REV. MOD. PHYS. **82**, 2365 (2010).
 - [16] J. Enders, *et al.*, PHYS. REV. LETT. **79**, 2010 (1997).
 - [17] E. Kwan, *et al.*, PHYS. REV. C**83**, 041601(R) (2011).
 - [18] S.L. Hammond, *et al.*, PHYS. REV. C**85**, 044302 (2012).
 - [19] J. Margraf, *et al.*, PHYS. REV. C**42**, 771 (1990).
 - [20] A.A. Kuliev, *et al.*, EUR. PHYS. J. A **43**, 313 (2010).
 - [21] M. Guttormsen, *et al.*, PHYS. REV. LETT. **109**, 162503 (2012).